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Roughness Analysis of Variously Polished Niobium Surfaces

Guilhem Ribeill and Charles Reece

ABSTRACT

Niobium superconducting radio frequency (SRF) cavities have gained widespread use in accelerator systems. It has been shown that surface roughness is a determining factor in the cavities' efficiency and maximum accelerating potential achievable through this technology. Irregularities in the surface can lead to spot heating, undesirable local electrical field enhancement and electron multipacting. Surface quality is typically ensured through the use of acid etching in a Buffered Chemical Polish (BCP) bath and electropolishing (EP). In this study, the effects of these techniques on surface morphology have been investigated in depth. The surface of niobium samples polished using different combinations of these techniques has been characterized through atomic force microscopy (AFM) and stylus profilometry across a range of length scales. The surface morphology was analyzed using spectral techniques to determine roughness and characteristic dimensions. Experimentation has shown that this method is a valuable tool that provides quantitative information about surface roughness at different length scales. It has demonstrated that light BCP pretreatment and lower electrolyte temperature favors a smoother electropolish. These results will allow for the design of a superior polishing process for niobium SRF cavities and therefore increased accelerator operating efficiency and power.

Introduction

Superconducting radio frequency (SRF) cavities are widely used to achieve high quality particle beams for nuclear physics research. Their usefulness rests mainly on their high quality factor, low power dissipation and potential for continuous wave operation. The most common material for the construction of these cavities is solid niobium, due to the availability of high-purity niobium and the ease of machining it into cavities [1,2]. Surface roughness has been shown to be a critical factor in determining the efficiency and maximum accelerating potential these cavities can achieve. The quality of the niobium surface directly impacts such undesirable effects such as field enhancement, the creation of 'hot spots' and electron multipacting [3]. The surface is typically polished using an acid etch Buffered Chemical Polish (BCP) consisting of HNO_3, HF and H_3PO_4 or EP in a HF/H_2SO_4 solution[4, 5].

Characterizing the surface morphology of the Niobium surfaces is a challenging task as it is unknown at what scale roughness is important to SRF performance. Pending such an analysis, the roughness of niobium must be looked at across a range of sizes and resolutions, which can only be accomplished using several different instruments. A method of combining the measurements from different instruments is therefore needed. A powerful tool for this kind of analysis is the power spectral density (PSD) [6]. From the PSD, one can easily obtain the characteristic length of surface features as well as the fractal dimension of the surface [7].

Sample Preparation

Chemical reagents used for polishing in this study were a BCP acid etch solution and an EP solution. The BCP solution was a 2:1:1 solution of 85% H_3PO_4, 49% HF and 70% HNO_3. The EP solution was a 1:9 mixture of 49% HF and 96% H_2SO_4. The samples that were used in this study were reactor-grade polycrystalline niobium slabs (CBMM, Sao Paulo, Brazil) with dimensions of 22 mm x 22 mm x 4 mm. All samples were mechanically polished using 1 μm grit prior to any chemical treatment. The sample used as a reference for an "untreated" sample was placed in a BCP acid etch for 5 minutes in order to remove any residual surface contamination. In this paper, the samples we refer to as having undergone a "Light BCP" treatment were placed in a BCP bath for 20 minutes. As measured by weight loss, the acid etch removed 30 μm. Those

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samples we refer to as having undergone "Heavy BCP" were placed in the BCP bath for 90 minutes which resulted in a 150 µm etch of the surface.

To electropolish the samples, the niobium sample and an aluminum counter electrode were held in place by Teflon clamps in the EP solution a distance of 100 mm apart. This setup was also used for taking EIS data with a mercury-amalgam reference electrode placed halfway between the niobium and counter-electrode. To prevent damage to the reference electrode, it was immersed in a solution of 1 N sulfuric acid, which was separated from the EP bath by a glass frit at the end of a Teflon tube. Samples having undergone no BCP treatment, as well as those having undergone light and heavy BCP treatment were subjected to 30 minutes of EP at 6 V, with electrolyte temperatures of 24°C and 30°C. These treatments removed approximately 100 µm from the surface, as measured using a stylus profilometer.

METHOD

Profilometry measurements were obtained with a stylus profilometer (KLA-Tencor: P-15) equipped with a tip with a diameter of two µm. The samples were scanned in two different regions with a scan size of 200 x 200 µm and 1000 x 1000 µm. The 200 µm scan was taken as an array of 101 traces with 401 points in each trace, and the 1000 µm as an array of 251 traces with 2501 points each. AFM measurements were performed using a commercial AFM (Digital Instruments: Nanoscope II) in tapping mode using silicon tips with a diameter of 10 nm. The samples were each scanned in three different regions with scan sizes of 20x20 µm, 50x50 µm, and 100x100 µm. The AFM images were captured as arrays of height values with 256x256 points.

There exist several measures of surface roughness. The typical measure is the root mean square roughness, defined by (1):

$$R_q = \sqrt{\frac{1}{n} \sum_{i,j} (z_{i,j} - \bar{z})^2} \tag{1}$$

Another way to characterize roughness is the Power Spectral Density (PSD). It can easily be computed as the square of the Fourier transform (2),

$$PSD(f) = \frac{1}{L} \left| \int_0^L dx z(x) e^{i2\pi fx} \right|^2 \tag{2}$$

or in the case of discrete data consisting of N points separated by Δx, equation (3),

$$PSD(m) = \frac{\Delta x}{N} \left| \sum_{n=0}^{N-1} z(n) e^{-i2\pi nm/N} \right|^2 \tag{3}$$

A more complete mathematical treatment can be found in Elson and Bennett [8]. The PSD represents the squared amplitude of surface features plotted against the spatial frequency of those features. This provides information about both the lateral and vertical size of features; data from different instruments can also be combined, taking care that the range of spatial frequencies over which the PSD is valid for a certain scan size is determined by the Nyquist limits. To simplify calculations, in this study the power spectral density for all traces in the fast scan direction were averaged to approximate the true 2D PSD. This assumption is valid in the case of a surface with isotropic roughness [9]. For all the samples, all the PSDs from a sample were averaged together and the PSDs

from the profilometer data were filtered using a 4-sample moving average in order to eliminate high-frequency noise.

RESULTS AND DISCUSSION

Figure 1 represents the 50 µm AFM data from the surface with five minutes of BCP and the surface that was treated with both heavy BCP and EP. The former shows very pronounced peaks and valleys, as well as a very non-uniform surface, whereas the latter is much smoother.

Figure 1. Superposition of typical scans from the surface having received 5 minutes of BCP, in blue, and the surface having received both heavy BCP and EP, in red, clearly showing the smoothing effect of these treatments. The small peaks in the heavy BCP/EP surface are likely due to instrument noise or surface contamination.
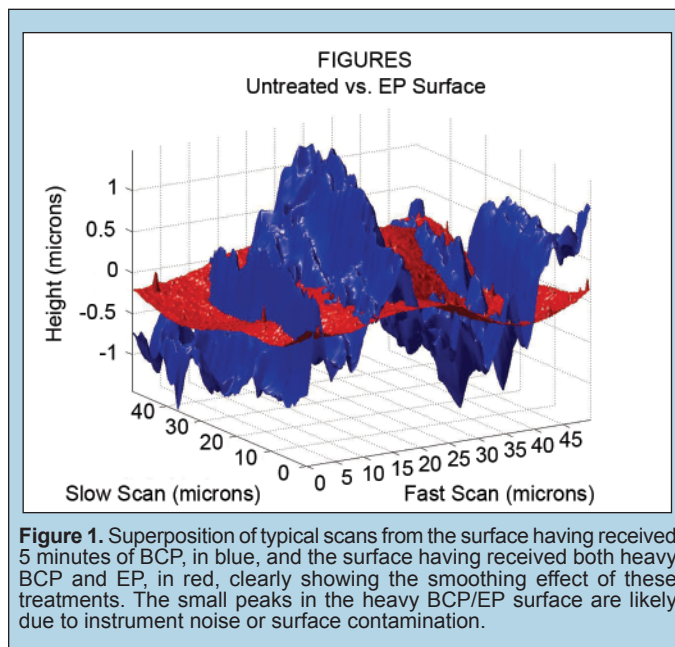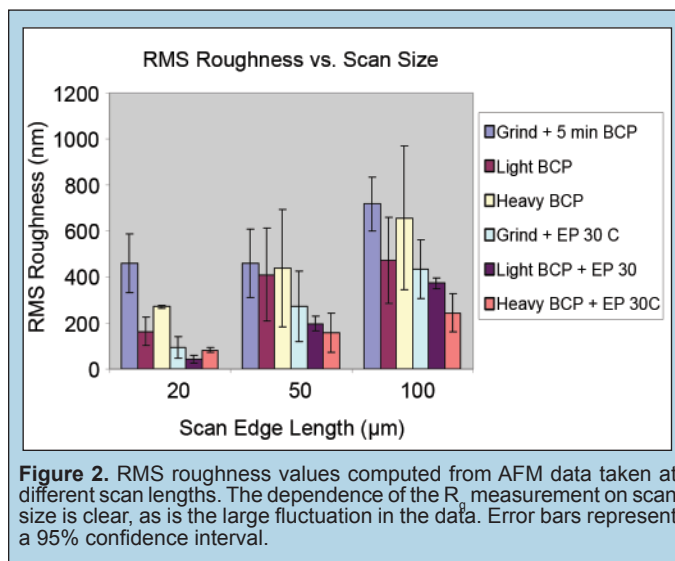
Figure 2 shows a plot of different R_q values computed from the data at all of the scan sizes. While R_q is a useful and widely used measure of surface roughness, it is dependent on the scan size and the particularities of the area being scanned. The data shows different values of R_q for each surface; at small scan lengths, light BCP + EP

Figure 2. RMS roughness values computed from AFM data taken at different scan lengths. The dependence of the R_q measurement on scan size is clear, as is the large fluctuation in the data. Error bars represent a 95% confidence interval.

is smoother than heavy BCP + EP, but it is rougher at large scan lengths. While this may indicate that heavy BCP followed by EP promotes smoothing only at large scales, large error bars associated with the R_q measurements, make it difficult to draw conclusions.

It is also difficult to compare roughness values taken using different instruments, especially when one considers that the range of frequencies that any one instrument can resolve is necessarily limited and unique to that instrument [6, 8]. Furthermore, since this roughness measurement considers only vertical information, it does not give any information about the lateral morphology of surface features.

In order to overcome these limitations, we have used the PSD, defined in equation 3, of the surface profile as a tool to combine measurements from different scales and different instruments.

Figure 3 shows the combined AFM and profilometer data from the sample with five minutes of BCP treatment. The measurements using the two techniques agree very closely over the range of frequencies that are common to both instruments. This shows that the PSD is a method which can combine information from different instruments in a consistent and useful manner. At the high-frequency end of the spectra, the slope of all the graphs shows a marked decrease and they begin to level off. This is likely due to the effects of limited resolution for the AFM, and the finite tip size for the profilometer data [10]. Of particular interest in this graph is the change of slope that occurs at a spatial frequency of approximately 0.04 μm. (From Fig.3 it is hard to tell the slope change around 0.04 μm^{-1}.) Changes in slope of the PSD are related to the inverse of the correlation length of the sample. The correlation length sets the scale at which surface features are similar [7, 9]. For this data, the correlation length is approximately 25 μm s, which is on the order of the size of niobium grains in the polycrystalline crystals studied.

For clarity, the rest of the graphs in this paper only show data from the 1000 μm profilometer scans and the 20 μm AFM scans. The differences between these scans and the others are minor, as can be seen in Fig. 3.
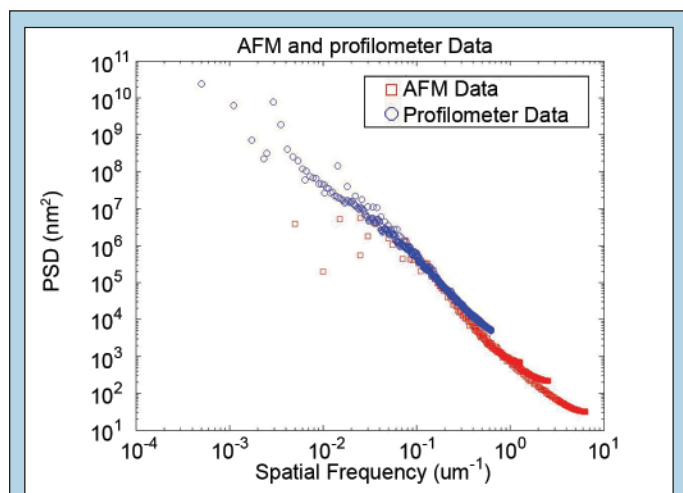
A comparison of the different methods of polishing a ground sample can be found in Figure 4. The smaller amplitude of the PSD for the electropolished sample supports the conclusion from R_q data that it is the technique that produces the smoothest surface. The slope change at 0.04 μm^{-1} from the ground sample is no longer visible, indicating that differences between grains have been greatly reduced, unlike with BCP. Furthermore, the greater slope of the PSD in the mid to high frequency region as compared to the other polishing techniques indicates that EP provides the best micropolish. The EP PSD also shows a change in slope at the smallest frequency, corresponding to EP producing a surface with the longest correlation length. This also supports the conclusion that EP produces the smoothest surface, since surface features are self-similar on larger scales. Interestingly, the light BCP treatment seems to produce a smaller PSD at low frequencies than the heavy BCP treatment, but the situation is reversed at high frequencies. Quite right, I took it out! This effect would be impossible to analyze if using only R_q as a measure of roughness; it indicates that a longer BCP treatment produces a better micropolish, but a rougher macropolish. This effect is likely due to the differential etching by BCP of grains with different crystallographic orientations [3]. As more material is etched, and the damage caused by the mechanical polish is removed, material from different grains is removed at different rates, leaving a rougher surface.
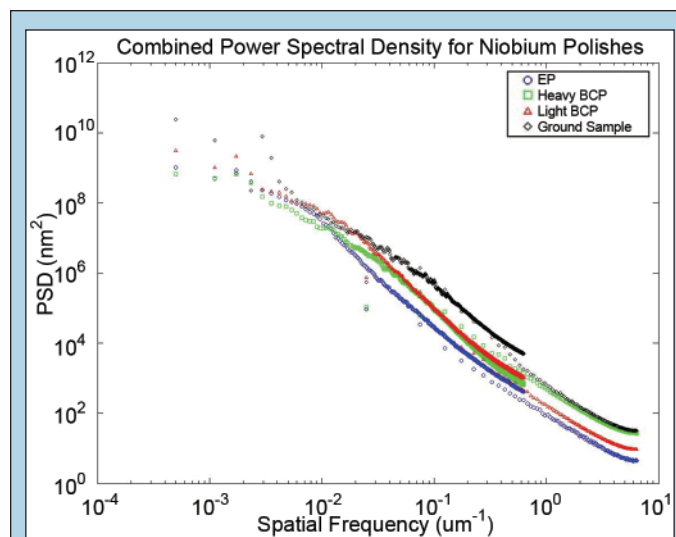

Figure 4. PSD spectra for the different polishing methods performed on a mechanically ground sample. Electropolishing was performed at 24°C.

Figure 5 compares the PSD of samples that have undergone different pretreatments before the electropolish. Light BCP again has the smallest amplitude, and the longest correlation length which corresponds to the smoothest surface. This result is not surprising in the context of the results previously discussed; a longer BCP treatment produces a rougher surface at large scales which has not been smoothed out by EP. Figure 6 presents the power spectra of ground samples treated with EP at two different electrolyte temperatures. The overlap of the two plots in the high-frequency region indicates that there is no significant difference in the quality of the polish at small scales. There is some evidence for a difference

Figure 3. Combined AFM and profilometer PSD from all scan sizes from the untreated sample.

at low frequencies, since the graphs take on a different shape. The longer correlation length of the 24°C sample, as indicated by the smaller inverse length at which the graph levels off, suggests that the lower temperature may cause a smoother finish of the sample, a conclusion also supported by the RMS roughness values, 84 nm for the 24°C niobium sample and 94 for the 30°C sample, at a scan size of 20 x 20 μm s. However, while suggestive, such a small temperature difference does not fully explore the effect of temperature on EP treatment and this merits further study.
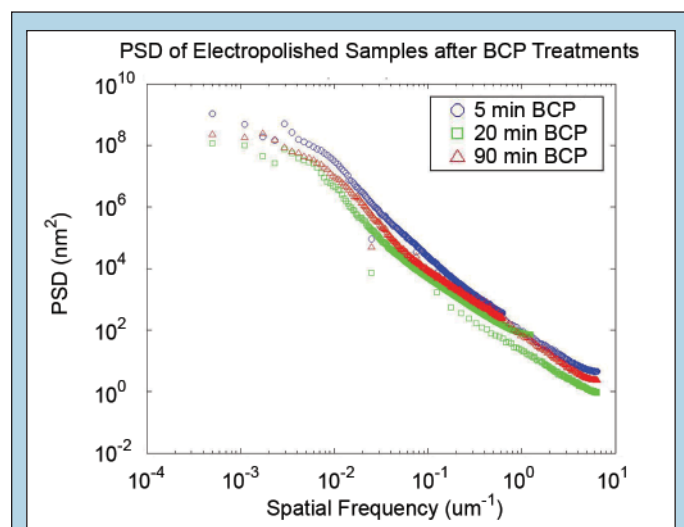
Figure 5. Power spectral density plots of niobium samples that have undergone different treatments before being electropolished.
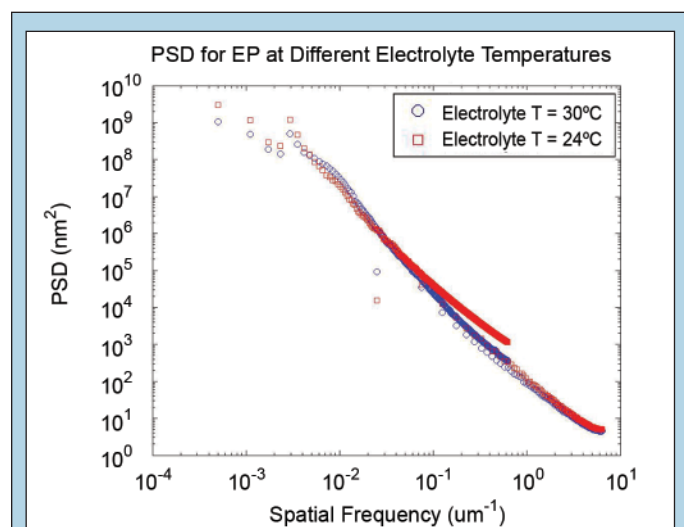
Figure 6. PSD graphs for EP of a ground sample at different electrolyte temperature.

CONCLUSION

We have demonstrated the use of spectral analysis techniques and, specifically, the power spectral density in quantitatively characterizing the surface roughness of niobium. This tool will therefore allow for improved monitoring and analysis in the production of SRF cavities. PSD curves can also be analyzed to determine fractal dimension, an alternative measure of surface roughness [7, 12]. In certain cases, this fractal dimension can be related to impedance spectra which show power-law dependence at low frequencies [13]. If this holds true in the case of niobium, it may be possible to use EIS to monitor surface roughness *in situ* during electropolishing.

The spectral technique has allowed us to make preliminary observations about the use of BCP and EP on niobium samples. We have conclusively shown that electropolishing produces the best surface polish, and is most effective when following a short BCP treatment. (Actually, in this manuscript, only one set of data (Fig. 2, scan size 20 × 20 μm) shows the sample with EP + light BCP gives best polishing. No other figures or PSD data demonstrate this point. So, more convincing data and explanation need be given.) Our data also suggests that a lower electrolyte temperature promotes a better EP at larger scales. However, our data is preliminary and merits much further study. The most important future work will be to determine at length scales roughness is most important to cavity performance. This information will allow for the development of a polishing process that optimizes the performance of SRF cavities and thereby accelerator systems.

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